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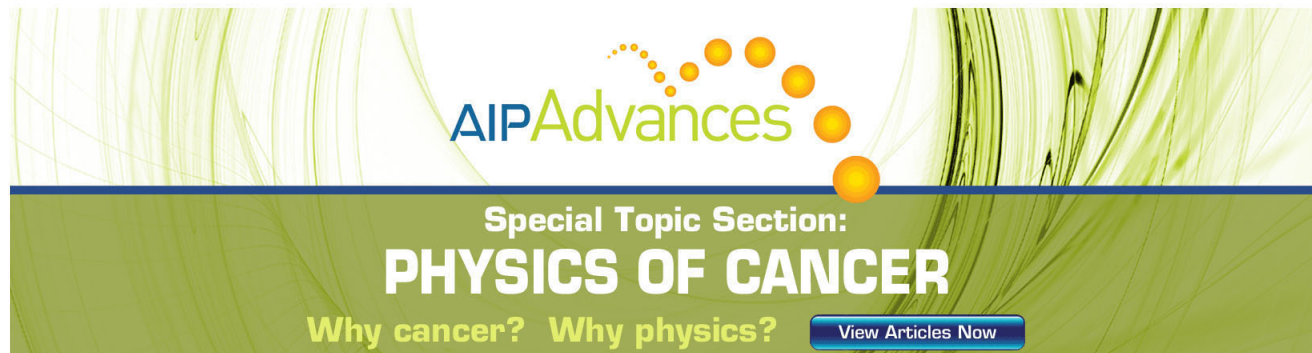
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Folded acoustic phonons in InAs-AlAs strained-layer superlattices

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Raman spectroscopy is used to characterize highly mismatched (7%) InAs-AlAs superlattices grown by atomic layer molecular beam epitaxy. In particular, folded acoustic modes are presented and compared with two different theoretical models (Rytov and linear chain). We find good agreement between theory and experiments. We estimate, with a simple model, the magnitude of the effect of the strain on the phonon frequency shifts.

Raman spectroscopy is a very useful tool to study lattice dynamics in bulk semiconductors. Because the light's wave vector is very small compared to lattice K vectors, most of the studies have been related to the easily observable optical phonon modes. In the case of superlattices where the bulk dispersion curves fold and modify, acoustic modes become also observable with a conventional Raman spectrometer. Consequently, from the position and intensity of the Raman peaks corresponding to folded modes, information on the superlattice quality can be achieved.

Different superlattices comprising lattice-matched (GaAs-AlAs)¹ and mismatched (GaAs-InGaAs, Si/GeSi)^{2,3} and even amorphous superlattices⁴ have shown those folding effects in their acoustic branches. The layered elastic continuum model of Rytov⁵ has been the most extensively used to describe the energy position of the folded acoustic phonons.

In this letter we present for the first time Raman scattering results of folded acoustic phonons in highly strained AlAs-InAs short-period superlattices (SLS) grown by atomic layer molecular beam epitaxy (ALMBE)⁶ on GaAs substrates. ALMBE is a novel modification of MBE in which group V effusion cell is pulsed in synchronism with the layer-by-layer growth sequence characteristic of III-V compounds.

Due to the forced two-dimensional (2D) growth mode induced by ALMBE, rough nucleation and growth typical of highly mismatched systems such as InAs on AlAs can be avoided. Substrate temperature and growth rate were 400 °C and 1 $\mu\text{m h}^{-1}$, respectively, for the present samples.

In this letter, experimental results in the acoustical mode region are discussed and compared with two different simple models: Rytov's model and the linear-chain model.⁷ Also, a correction on both models which takes into account internal stresses is introduced.

Rytov's model is based on a quasilinearity of the phonon dispersion branch in the low-frequency range, and takes the superlattice as an elastic continuum composed by the repetition of two layers of materials 1 and 2. The dispersion curve along the superlattice growth axis can be expressed as

$$\cos(q_z d) = \cos\left(\frac{wd_1}{V_1}\right) \cos\left(\frac{wd_2}{V_2}\right) - \left(\frac{\theta^2 + 1}{2\theta}\right) \times \sin\left(\frac{wd_1}{V_1}\right) \sin\left(\frac{wd_2}{V_2}\right), \quad (1)$$

$$\theta = \frac{\rho_2}{\rho_1} \frac{V_2}{V_1},$$

where q_z is the superlattice wave vector in the growth direction (z), d_1 and d_2 are the thicknesses of layers 1 and 2, $d = d_1 + d_2$ is the superlattice period, and ρ_1, ρ_2, V_1, V_2 are the densities and sound velocities of the two constituent materials.

When the period of the superlattice is small enough a linear approximation for the dispersion curve becomes inappropriate. A linear-chain model is expected to describe better the behavior of the phonon dispersion when not in the linear region. In such a model, the dispersion relation for the phonons in the superlattice is given solving Eqs. (2), (3), and (4).

$$\cos(\alpha a_1) = [(m_1 w^2 - 2K_1)(m_2 w^2 - 2K_1) - 2K_1^2] / 2K_1^2 \quad (2)$$

$$\cos(\beta a_2) = [(m_1 w^2 - 2K_2)(m_2 w^2 - 2K_2) - 2K_2^2] / 2K_2^2 \quad (3)$$

$$\cos(qd) = \cos(\alpha d_1) \cos(\beta d_2) + C \sin(\alpha d_1) \sin(\beta d_2), \quad (4)$$

being

$$C = -\frac{1}{2} \left(\frac{m_2 w^2 - 2K_2}{m_2 w^2 - 2K_1} \frac{[1 + \cos(\alpha a_1)][1 - \cos(\beta d_2)]}{\sin(\alpha a_1) \sin(\beta a_2)} + \frac{m_2 w^2 - 2K_1}{m_2 w^2 - 2K_2} \frac{[1 - \cos(\alpha a_1)][1 + \cos(\beta a_2)]}{\sin(\alpha a_1) \sin(\beta a_2)} \right),$$

where m_2 is the mass of the common As atom, m_1 and m_1' are the Al and In atom masses, a_1 and a_2 are the half-lattice constant of materials 1 and 2, and α and β are the complex wave vectors for the bulk materials 1 and 2. The force constants are noted in the expression as K_1 and K_2 .

As it is well known, the linear-chain model does not reproduce the whole phonon dispersion curves of the bulk material. In order to overcome this discrepancy we have allowed force constants K_j to vary according to the following law: at the Γ point we have chosen a value of K_j that could fit V_{001} , and at the X point a value of K_j that fits the frequency of the longitudinal acoustic LA(X) phonon. In between those two extreme points K_j varies linearly.

Raman spectra were obtained at room temperature in the backscattering configuration. The scattered light was analyzed by means of a computer-controlled 0.85 m double monochromator.

The geometrical parameters of the superlattices, determined by x-ray techniques, reflection high-energy electron diffraction (RHEED) during growth, and tested by analysis of the Al and In content (energy dispersive analysis of x-ray

TABLE I. Sample structures.

Sample	Period ($d_1 + d_2$, Å)	InAs thickness (d_1 , Å)	AlAs thickness (d_2 , Å)
S1	11.9	9.1	2.8
S2	29.7	21.2	8.5
S3	38.8	30.3	8.5
S4	62.6	48.5	14.2

EDAX), are displayed in Table I. Periods vary over a range from 12 to 62.6 Å.

In Fig. 1, Raman spectra are displayed for the four samples S1, S2, S3, and S4.

Raman spectra for samples S3 and S4 (shown in Fig. 1) show a well-resolved first ($m = 1$) doublet with a frequency

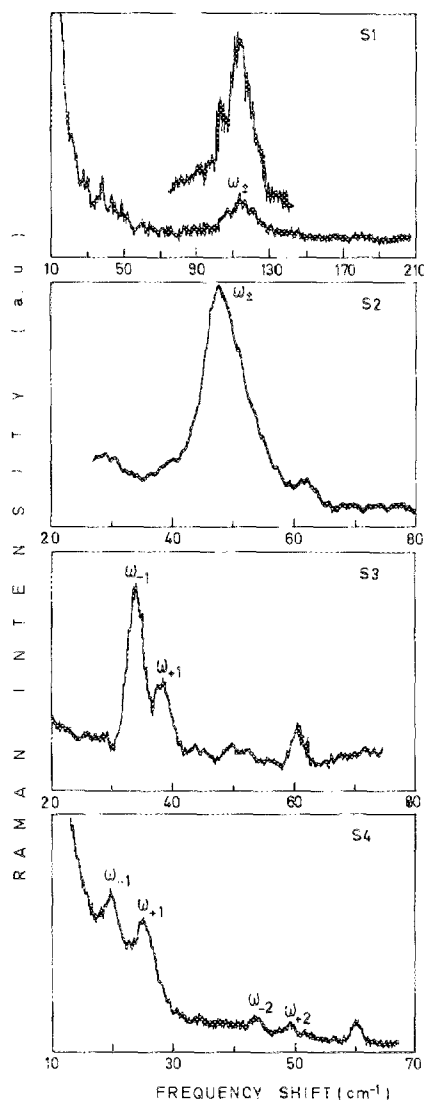


FIG. 1. Raman spectra from samples S1, S2, S3, and S4 in Table I, recorded at room temperature. The $m = 1$ and $m = 2$ doublets are clearly resolved in S4 while in S1, S2, and S3 only the $m = 1$ doublet is observed. See Table II for the values of their peak frequencies. The peak at 60 cm^{-1} is associated to scattering by transversal acoustic TA phonons [this frequency corresponds to the InAs TA(X) phonons].

distance between W_{-1} and W_{+1} of 4.5 and 5 cm^{-1} , respectively. In sample S4 the $m = 2$ doublet is also observed.

S1, the sample with the shortest period, presents a structured broad peak that could be attributed to the first LA doublet with a frequency distance between W_{-1} and W_{+1} of 9 cm^{-1} .

In sample S2, a single peak with frequency value noted as W_{\pm} in Table II is observed. The nonobservation of a doublet of splitting might be related to lower interfacial quality in this particular sample.

Parameters used in the Rytov and linear-chain model calculations were V_1 (AlAs) = $565\,403 \text{ cm/s}$, V_2 (InAs) = $383\,608 \text{ cm/s}$, $\rho_1 = 3.76 \text{ g/cm}^3$, $\rho_2 = 5.66 \text{ g/cm}^3$, and the SLS refractive index $n_{\text{SL}} = 4.3$. We have taken the equivalent alloy of our superlattices and supposed a linear variation of the refractive index n between the indexes of these two materials. Table II shows all experimental and theoretical data (Rytov and linear-chain calculations) of the doublets obtained from samples S1 to S4.

As mentioned before, Rytov's model is suitable for "long enough" superlattice periods, where the experimental q_z lies on the linear region of the phonon dispersion curve. Good agreement between experimental results and Rytov's model is found for the three samples with longer period S2, S3, and S4. Also the separation between W_{-1} and W_{+1} is well described. In these samples the linear-chain model, as presented here, agrees well with the experimental data also.

Rytov's model gives a W_{-j} to W_{+j} constant separation independently of the superlattice period; for example, in sample S1, Rytov's model gives $W_{+1} = 5 \text{ cm}^{-1}$, while the linear-chain model gives 10 cm^{-1} , which agrees better with the suggestion made before associating the first LA doublet to the "structured" broad peak in sample S1.

As can be seen, no substantial deviations are observed from the calculated values using the Rytov and linear-chain models, and the experimental results, which means that both models are still good approximations in describing our large mismatched InAs-AlAs superlattices.

Due to the fact that the in-plane lattice parameter has a constant value between those of InAs and AlAs, the InAs layers are under biaxial compression whereas the AlAs layers are under biaxial extension. In order to have a rough

TABLE II. Experimental and theoretical frequencies for the folded LA phonons of the superlattices (cm^{-1}).

Sample	Experimental frequency	Rytov model	L.C. ^a model	Rytov + stress	L.C. ^a + stress
S1	ω_{-1}	110.0	113.5	107.0	117.5
	ω_{+1}	119.0	118.5	117.0	123.5
S2	ω_{\pm}	47.5	45.0	44.5	46.0
			49.5	50.0	51.0
S3	ω_{-1}	33.7	33.0	33.0	34.0
	ω_{+1}	38.2	37.5	38.0	39.0
	ω_{-1}	19.6	19.5	19.5	20.0
	ω_{+1}	24.6	24.0	24.5	25.0
S4	ω_{-2}	43.5	41.5	41.5	43.0
	ω_{+2}	49.0	46.0	46.5	48.0

^a L.C.-linear chain.

estimation of the magnitude of these effects on the phonon dispersion curves of the superlattice, we have proceeded as follows: the modification of the sound velocity and the LA(X) phonon due to hydrostatic pressure is related to the shift of the longitudinal optical (LO) phonon by

$$\frac{\Delta v}{v} = \frac{\gamma_{LA}(\Gamma)}{\gamma_{LO}(\Gamma)} \frac{\Delta w}{w}, \quad (5)$$

$$\frac{\Delta w_{LA}(x)}{w_{LA}(x)} = \frac{\gamma_{LA}(x)}{\gamma_{LO}(\Gamma)} \frac{\Delta w_{LO}(\Gamma)}{w_{LO}(\Gamma)}, \quad (6)$$

where γ_{LA} and γ_{LO} are the Grüneisen parameters for the LA and LO phonons, respectively. Up to our knowledge the only reported experimental values for these parameters are those of the LO(Γ) phonon in InAs.⁸ So we have taken the theoretical values for the Grüneisen parameters of AlAs given in Ref. 9 and a value of $\gamma_{LA} = 1.34$ for the acoustical phonon branch of InAs.¹⁰ (No modification of the Grüneisen parameters along the dispersion curves has been included in the calculations.)

Using the measured strain shift of the optical phonon¹¹ we can deduce the new sound velocities and the energy shift of the LA phonons. The results of such a modification for both models (Rytov and linear chain) are shown in Table II.

Looking at the results in Table II, it is obvious that the effect on the LA phonons of the 7% mismatch does not seem to be important.

Summarizing, we have presented results on folded acoustic phonons on InAs-AlAs highly strained-layer superlattices. The results are well described by both Rytov's and the linear-chain model. Corrections of the sound velocity due to the enormous strain level in the layers give rise to very small energy shifts in the superlattice acoustic modes in agreement with experimental observations of folded acoustic LA phonons.

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